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SHEAR MODELING: THERMOELASTICITY AT HIGH TEMPERATURE AND PRESSURE FOR TANTALUM

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ABSTRACT- For large-scale constitutive strength models the shear modulus is typically assumed to be linearly dependent on temperature. However, for materials compressed beyond the Hugoniot or in regimes where there is very little experimental data, accurate and validated models must be used. To this end, we present here a new methodology that fully accounts for electron- and ion-thermal contributions to the elastic moduli over broad ranges of temperature (<20,000 K) and pressure (<10 Mbar). In this approach, the full potential linear muffin-tin orbital (FP-LMTO) method for the cold and electron-thermal contributions is closely coupled with ion-thermal contributions. For the latter two separate approaches are used. In one approach, the quasi-harmonic, ion-thermal contribution is obtained through a Brillouin zone sum of strain derivatives of the phonons, and in the other a full anharmonic ion-thermal contribution is obtained directly through Monte Carlo (MC) canonical distribution averages of strain derivatives on the multi-ion potential itself. Both approaches use quantum-based interatomic potentials derived from model generalized pseudopotential theory (MGPT). For tantalum, the resulting elastic moduli are compared to available ultrasonic measurements and diamond-anvil-cell compression experiments. Over the range of temperature and pressure considered, the results are then used in a polycrystalline averaging for the shear modulus to assess the linear temperature dependence for Ta.

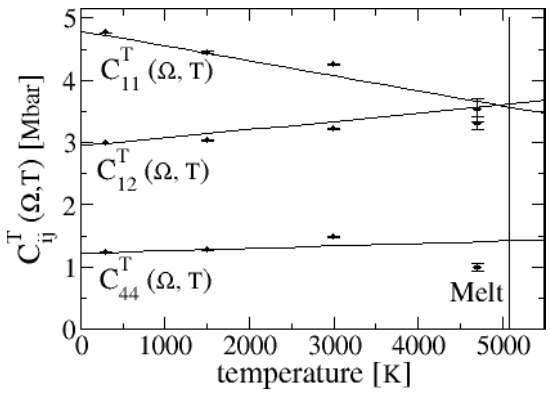

Fig. 1 A comparison between QH (lines) and AH (points) ion thermal components to the C_{ijkl}^T at Ω=102.2 a.u.^{3} indicating minimal AH effects. The pressure varies from 0.5 to 0.7 Mbar nearing T_m.

Typically for large-scale computations constitutive strength models contain a polycrystalline shear modulus that has an assumed linear temperature dependence (Steinberg [1980], Preston [2003]). To assess the validity of this assumption, we present a new methodology and results for single-crystal elastic moduli and the polycrystalline-averaged shear modulus G for the high temperature and pressure phase diagram of tantalum up to 20000 K and 10 Mbar, where both ionic and electronic thermal contributions are fully treated. Previous work on the pressure dependence of the cold shear modulus (T=0K) indicates a non-linear pressure dependence beyond 5.5 Mbar (Söderlind [1998]). Despite this pressure behavior in this case for Ta, the temperature dependence of G at a fixed volume is linear up to melt due to anharmonic effects being negligible for this metal.

For high temperatures (300 K ≤ T ≤ T_{melt}) and high pressures (P < 10 Mbar), we assume that the electron-phonon coupling is negligible for a metal and write the Helmholtz free

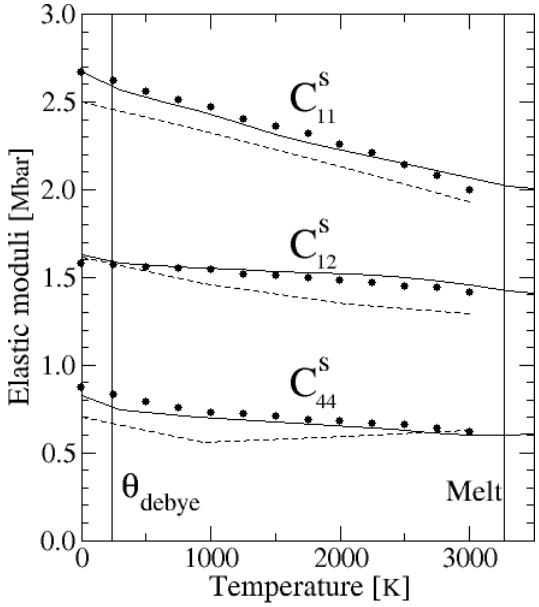

Fig. 2 The fully calculated C^S_{ijkl} (solid lines) at P=0 compared to experiment(Walker[1980]) (circles). The dashed lines are the calculations from Gülseren[2002].

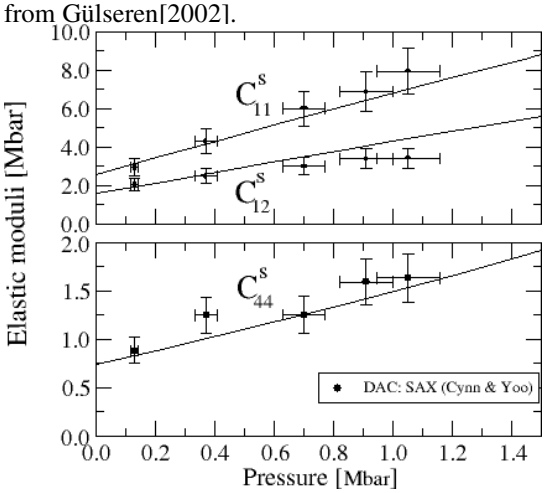

Fig. 3 The fully calculated C^S_{ijkl} (lines) at T=300 K as a function of pressure compared to DAC: SAX data(Cynn[2002]).

energy, $F(\Omega,T) = \Phi_o(\Omega,T=0) + F_e(\Omega,T) + F_H(\Omega,T) + F_A(\Omega,T)$, where $\Phi_o(\Omega,T=0)$ is the total energy of the electronic ground state, i.e. the frozen lattice, $F_e(\Omega,T)$ contains the electron-thermal contribution, $F_H(\Omega,T)$ holds the ion-thermal contribution, $F_A(\Omega,T)$ has the anharmonic(AH) contributions. The specific volume Ω is the volume per atom. With this and through the definition of the isothermal elastic moduli $C^T_{ijkl}(\Omega,T) = \partial^2 F(\Omega,T) \big/ \partial\eta_{ij}\partial\eta_{kl}\big|_{\eta'}$, where η' indicates that all other strains are held fixed, the individual contributions to the elastic moduli are obtained as $C^T_{ijkl}(\Omega,T) = C^o_{ijkl} + C^e_{ijkl} + C^{ion}_{ijkl}$. For the C^e_{ijkl} term, temperature is incorporated into F^e through a broadening of the electron density of states via the Fermi-Dirac distribution. To calculate this term, the full potential, linear muffin-tin orbital (FP-LMTO) electronic structure method is used (Söderlind[1998]).

For the $C^{ion}_{ijkl}(\Omega,T)$ contribution, we have implemented two different calculations to assess AH effects: one within the quasi-harmonic(QH) phonon approximation and one that is fully AH. Specifically for the QH method, following Wallace[1998], the Helmholtz free energy for the lattice is written as a Brillouin zone and branch sum of the phonon frequencies. Therefore, to obtain $C^{ion}_{ijkl}(\Omega,T)$ strain derivatives of F^{ion} lead to a summation over the Brillouin zone of strain derivatives of the phonon frequencies. To compute the fully AH lattice contribution to the elastic moduli, we have extended previous MC work (Greeff[1999]), where the strain derivatives of the partition function are taken while accounting for periodic boundary conditions. This leads to a canonical assemble average of these derivatives evaluated at thermodynamic equilibrium for a given Ω and T via a standard Metropolis, MC algorithm. Two to three runs for each (Ω,T) point were performed with a run lasting at least 1.5E6 MC-steps. In both ion-thermal methods, we have used a quantum derived, multi-ion potential for Ta from the model generalized pseudopotential theory(MGPT) (Moriarty[1990]).

We first establish that the AH effects are minimal for Ta over its phase diagram in Fig.1 showing typical behavior for only the ion-thermal component $C_{ijkl}^{ion}(\Omega,T)$ calculated via the above QH and AH methods for a given volume. Other volumes also indicate little deviation from linear temperature dependence for $C_{ijkl}^{ion}(\Omega,T)$ up to 10 Mbar. With negligible AH effects present for the Ta system, full calculations with electron- plus QH ion-thermal contributions for the adiabatic moduli C_{ijkl}^{S} obtained from $C_{ijkl}^{T}(\Omega,T)$ are compared against experimental data in Figs. 2 and 3. We have found that it is necessary to include both the electron- and ion-thermal components of the C_{ijkl}^{S}, since each component is of similar magnitude, roughly 0.1 Mbar at T=2500 K and P=0. In Fig. 4 the G(Ω,T) obtained by a Voigt average of the $C_{ijkl}^{T}(\Omega,T)$ for fixed volumes are shown. The overall behavior is linear in temperature for each Ω with fluctuations about a linear fit of <15%, which in turn indicates the band of confidence for G(Ω,T) in constitutive models for this particular case of Ta. Such linear temperature dependence is expected to break down in more AH systems like Mo, since the ion component becomes proportional to higher order temperature terms, especially at the system approaches its melt.

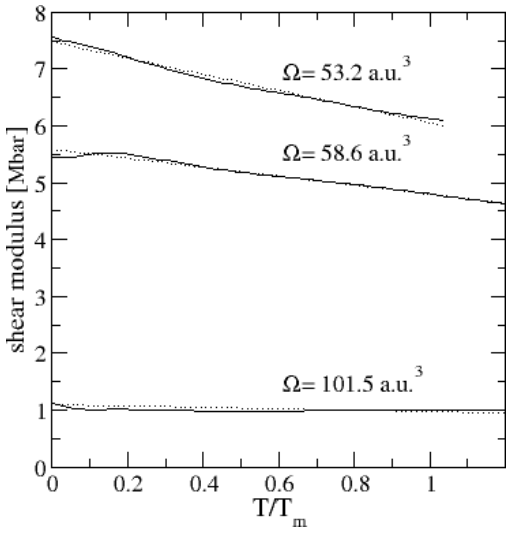

Fig. 4 The Voigt averaged G(Ω,T)(solid line) as a function of temperature scaled by the T_{melt} for fixed volume in comparison with linear fits (dotted line).

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